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Advances in Hydrogen Management for High Strength Steel

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ABSTRACT

Recent advances in hydrogen management for high strength steel welds are reviewed and show promise for higher performance low hydrogen welding consumables.

INTRODUCTION

High strength low alloy (HSLA) steels are known to be susceptible to hydrogen cracking. Also, the hydrogen assisted cracking in HSLA steel welds is considered to take place when all the necessary conditions for cracking are satisfied simultaneously. These conditions include the combination of unacceptable diffusible hydrogen content, high tensile stress, high hardness or a susceptible microstructure, and a temperature ranging between –50 and 100 °C [1, 2, 3, 4, 5]. One of the main practices to make a HAC resisting high strength steel weldment is to reduce the amount of diffusible hydrogen. Another common practice is the pre- or post- weld heat treatment. However, heat treatments are cost intensive and, in some critical cases, not effective. Alternative methods based on metallurgical principles have been studied and offer both for technological and economic merits.

This paper focuses on the recent advances in hydrogen management in high strength steel, particularly on three strategies. First of all, proper selection of weld metal martensite (ferrite) start temperature is introduced. Second, Irreversible weld metal hydrogen traps, such as yttrium addition, have been demonstrated to be effective in managing diffusible hydrogen content, and thus the susceptibility for hydrogen assisted cracking. The influence of the welding parameters on yttrium transferability across the welding arc and on hydrogen trapping behavior in the weld deposition is presented to assist in transferring this advancement to welding practice. Finally, the use of fluoride additions to the welding consumables also has been shown to effectively reduce the weld metal hydrogen content. This paper also discussed about the role of retained austenite in the higher strength steel weld metal in storing and supplying diffusible hydrogen in the weld deposit and its behavior during changes in service temperature is reported.

I. PROPER SELECTION OF WELD METAL MARTENSITE START TEMPERATURE.

The steel weld hydrogen content is dependent on both the hydrogen source and the ability of the weldment to transport hydrogen from the weld metal to the heat affected zone. The transport aspect becomes important because hydrogen atoms have to migrate across two different phases during cooling, mainly austenite and ferrite (martensite). Austenite (FCC) has high hydrogen solubility but low diffusion rate of hydrogen, which in contrast to ferrite and martensite that have orders of magnitude higher hydrogen diffusion coefficients than austenite [6]. As a result of the different hydrogen diffusivities between the two phases, a non-uniform distribution of hydrogen can result across the weldment depending on austenite decomposition behavior of the alloys [7,8]. Evidence of this resulting localized hydrogen distribution can be seen in the laser induced breakdown spectroscopy data for hydrogen spectral emission scans across a weldment as shown in Figure 1 [9]. The apparent localization of hydrogen in this figure may due to the effect of residual stress associated with the weld fusion line, which is known to promote attraction for hydrogen [1]. The effect of hydrogen damage is magnified when the location of the susceptible microstructures (martensite) overlaps the local high hydrogen content.

Proper alloying elements and their contents in the consumable are to be used to insure maximum hydrogen transport away from the weldment (weld metal and HAZ) during the welding thermal cycle. As an alloying dependent parameter, the martensite start temperature (M_s) can be used as an indicator for effective transport of hydrogen. A large difference between the M_s (weld metal) and M_s (base metal) will indicate difficulties in hydrogen transport in the weldment and the tendency for high localization in hydrogen contents. Thus the M_s is a measure of both the microstructure evolution and the ability to have a combination of microstructures suitable for rapid hydrogen transport [7, 8, 10, 11].

Granjon [7] introduced a conceptual model that describes how the austenite-ferrite (or austenite-martensite) phase transformation in steel weldment affects the resulting hydrogen distribution. Two cases are illustrated in Figure 2. When the austenite-martensite transformation in the fusion zone (weld metal) occurs at a higher temperature than the heat affected zone diffusible hydrogen will segregate in the heat affected zone just under the fusion line. This HAZ region is often the location of underbead cracking in high strength steel weldments. On the other hand, when the martensite transformation in the heat affected zone occurs at a higher temperature than in the fusion zone, it is possible that excess hydrogen may accumulate in the weld metal. This situation could promote weld metal hydrogen cracking or micro-fissuring.
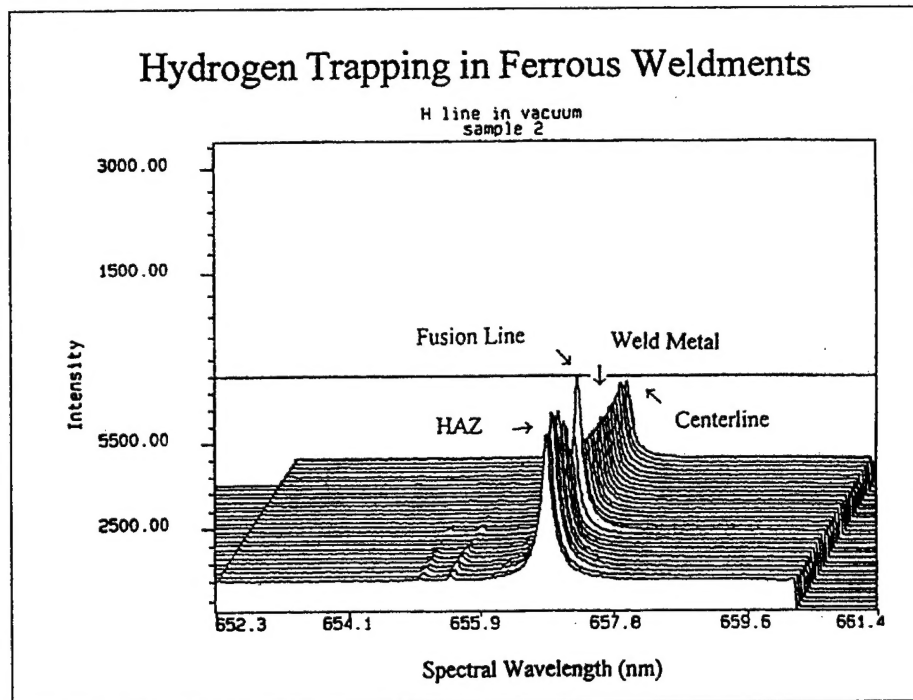
Figure 1. Non-uniform distribution of hydrogen across the center line of a weldment. Intensities of the hydrogen spectral emission are proportional to the hydrogen concentration [9].
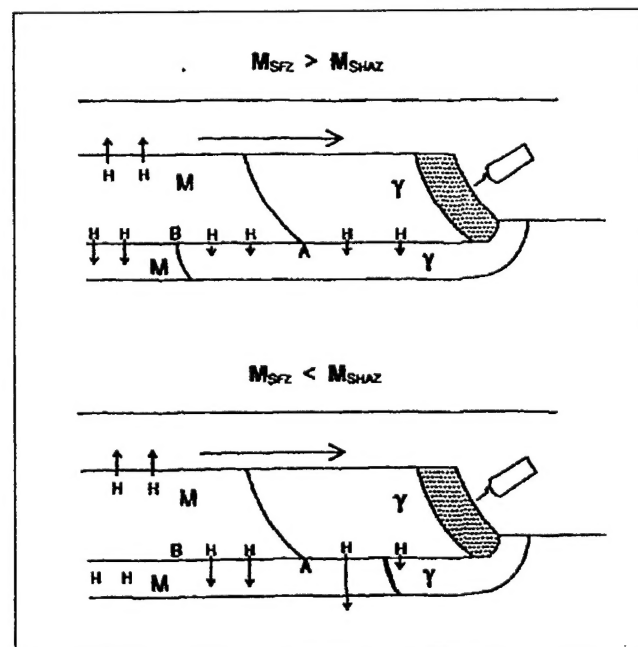

Figure 2. Illustration of hydrogen diffusion at different martensite start temperature for weldment and base metal [7].

CASE 1: WELD METAL WITH LOWER M_s TEMPERATURE THAN THE HAZ.

This case can be described as a situation where the weld metal is overmatched with respect to the base metal. The weld metal with higher alloying content exhibits a higher strength than the base metal, hence its martensite start temperature is depressed to below that of the base metal. While the austenite in the HAZ has begun transforming, the austenite in the weld metal remains unchanged. At this point, the HAZ immediately adjacent to the fusion zone will transport hydrogen at a higher rate than that in the weld metal. If the HAZ martensite start temperature (M_s) is sufficiently high, hydrogen will be able to migrate a significant distance into the parent metal. Especially if the transformation occurs at a moderately elevated temperature, the situation described can potentially reduce the localized hydrogen content in the hard microstructure adjacent to the fusion line, thus reducing the hydrogen cracking susceptibility. However, the hydrogen transport cannot proceed extensively until the weld metal transforms because austenite has the ability of storing high hydrogen contents but can not transport it fast enough to the fusion line. If the weld metal M_s temperature is too low, then the hydrogen transport from the weld metal is limited. Very little hydrogen can reach the heat affected zone adjacent to the fusion line that may eventually lead to weld metal cracking.

CASE 2: HAZ WITH LOWER M_s TEMPERATURE THAN THE WELD METAL.

This case can be described as a situation where the weld metal is undermatched with respect to the base metal. Since the heat affected zone transforms from austenite to ferrite/martensite at lower temperatures and at a later time than the weld metal, the HAZ becomes an austenite diffusion barrier for hydrogen transport. A high hydrogen accumulation in the heat affected zone adjacent to the fusion line results. This situation promotes underbead hydrogen cracking.

To evaluate the ability of using the martensite start temperature as a hydrogen cracking index, the diffusible hydrogen content was plotted as a function of the calculated weld metal martensite start temperature for welds made on the same high strength low alloy steel (Figure 3) [12]. Figure 3 illustrates a demarcation line between the cracked and the uncracked weldments.

From the application of the M_s temperature, it is possible to obtain a ΔM_s expression, and the sign and magnitude of this ΔM_s expression will better describe the hydrogen diffusion behavior:

$$\Delta M_s = M_{s(WM)} - M_{s(BM)}$$

If $\Delta M_s < 0$, hydrogen accumulation will be in the weld metal. If $\Delta M_s > 0$, hydrogen accumulation in the HAZ is possible and underbead cracking may occur. Selection of the alloy additions has to be determined to achieve only a slightly higher martensite start temperature of the weld metal than that of the HAZ, for maximum hydrogen transport to the base metal. In addition, the absolute martensite start temperatures of the weld metal and the HAZ should be high enough to facilitate rapid hydrogen transport in the martensite phase.
Since hydrogen is also known to migrate to regions of high residual tensile strength which

most often in the heat affective zone adjacent to the fusion line, the use irreversible traps may sufficiently immobilize the hydrogen to prevent delayed underbead cracking.
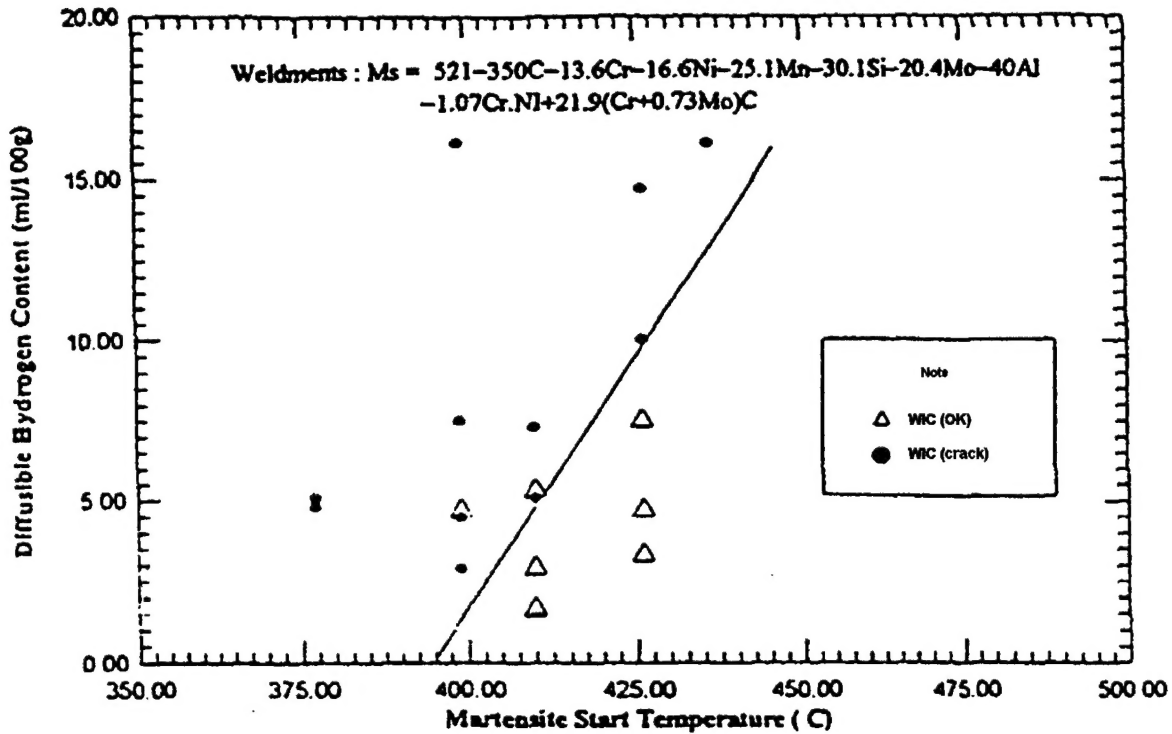

Figure 3. Conceptual illustration of cracking prediction [11].

II. DIFFUSIBLE HYDROGEN CONTROL BY IRREVERSIBLE HYDROGEN TRAPS

II-1 Rare Earth Metal Additions to the Weld Metal

It is known that inclusions in steel which contain elements on the left side of the transition metal series or rare earth elements have interfaces at which hydrogen is allowed to spend long resident times. Elemental additions, which allow for extremely long interfacial resident times, are classified as irreversible hydrogen traps. These weld metal irreversible hydrogen traps binds up the hydrogen at high temperatures and potentially reduces the diffusible hydrogen content [13, 14, 15].

The role of hydrogen trapping, especially strong traps, in high strength steel welding was investigated by adding selected rare earth metals to the weld metal. These additions were made during gas metal arc (GMA) welding using metal cored wires. The selected rare earth metals were neodymium and yttrium, which were inserted into the metal cored GMA-wire consumables in powder form as iron-neodymium or iron-yttrium intermetallic compounds. The powder size selected for the wire manufacturing was between -35 to -70 mesh.

Diffusible hydrogen measurements using ferro-yttrium additions to the metal cored wire are shown for two levels (1000 and 2000 ppm) of addition in Figure 4 [13]. This set of measurements also utilizes welding parameters which yields 1.5 kJ/mm and an argon-0.1 pct hydrogen shielding gas. Figure 4 clearly shows the direct proportionality of diffusible hydrogen reduction with higher amounts of yttrium. Almost 90 pct. diffusible hydrogen reduction can be achieved when welding with the wires that yield 2000 ppm weld metal yttrium. These results implies that addition of yttrium can be tailored to achieve a specific target value of reduction of the diffusible hydrogen content, as long as the other aspects of mechanical integrity of the weld joint are not severely affected.
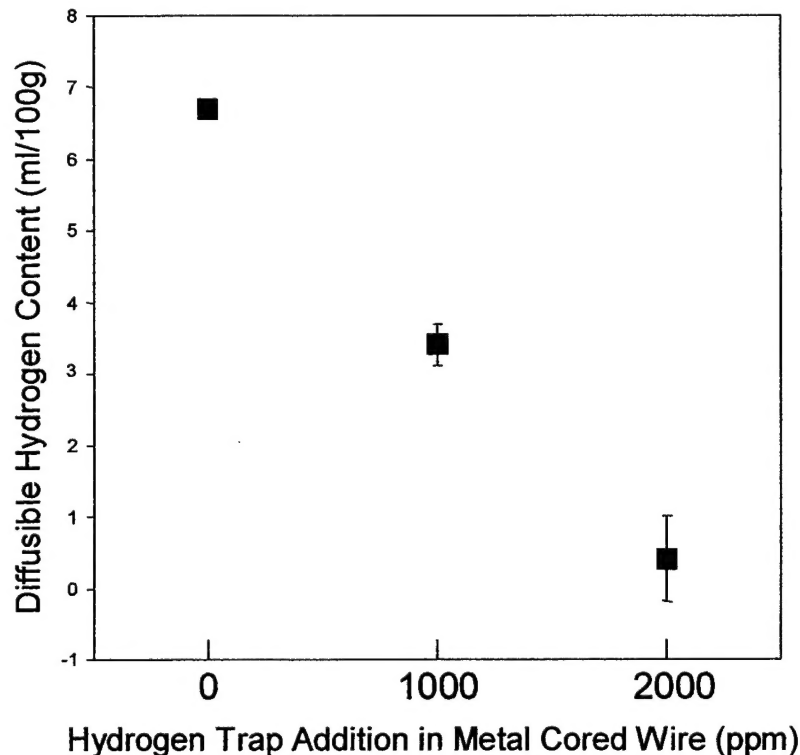
Fig. 4. Diffusible hydrogen content of HSLA steel weld sample GMA welded with mild steel formulated metal cored wires containing ferro-yttrium additions and with 0.1 pct H_2 in argon shielding gas [13].

Diffusible hydrogen measurements have effectively demonstrated the potential use of a hydrogen trapping concept in the hydrogen management of high strength steel welding. However, this measurement can not distinguish the performance of a strong trap site from a weak trap. The ice water quenching procedure stipulated by the AWS specification does not enable one to evaluate the hydrogen trapping performance of individual trap sites in the weld sample, especially above the critical hydrogen cracking temperature (100 $^\circ$C). Any kind of trap will perform well in such a condition. The accelerated reduction of diffusible hydrogen that is expected from the use of a strong trap can be evaluated by the hydrogen thermal desorption analysis. This measurement technique analyzes the release of hydrogen from various trap sites at characteristic temperatures during a constant heating rate of the weld

samples. The methodology applied in this analysis provides an easy way to evaluate a specific trap without excessive interference from other traps coexisting in the weld samples. Hydrogen thermal desorption analysis was used to show that the reduction is directly associated with the hydrogen trapping of the rare earth metal additions in the weld metal. It also served the purpose of showing the strength (binding energy) of the trap sites of interest, as a direct indication of its capacity to provide accelerated reduction of the diffusible hydrogen.

The hydrogen thermal desorption of yttrium containing high purity iron weld metal is shown in Figure 5.a., while that for HSLA 100 steel weld metal is in Figure 5.b [13]. Table 1 reports the chemical composition of HSLA steel and Iron used in this investigation. Also shown in these figures are hydrogen desorption of reference samples, which did not contain yttrium. Both hydrogen thermal desorptions were obtained after the measurement of diffusible hydrogen. They show the presence of hydrogen desorption with peaks at 600 ^{o}C and at 800 ^{o}C at 4°C/min (550 and 750 ^{o}C at 3°C/min). These desorption peaks indicate the presence of two kinds of strong trap sites in the weld metal. These desorption peaks also show that the reduction of diffusible hydrogen occurs because part of the total hydrogen was partitioned to the trap sites. The data suggests that on cooling, the yttrium containing weld deposits traps significant amount of the hydrogen at high temperature leaving the remaining hydrogen for the reversible traps and lattice sites. Identification of the trap sites present in the weld samples with electron microscopy and X-ray diffraction analysis indicates that the 600 ^{o}C desorption peak originated from Y_2O_3 inclusions and the 800 ^{o}C peak from Fe_2Y second phase.

Table I- Chemical Composition of HSLA Steel and Armco Iron Base Metal the Each Sample (wt-%)

	C	Mn	P	Si	Cr	Ni	Mo	Cu	Al	V	N	Ti
Armco Iron	0.004	1.19	0.009	0.06	0.03	0.007	0.003	0.014	0.006	<0.002	0.004	<0.002
HSLA Steel	0.06	0.83	0.01	0.35	0.57	3.42	0.62	1.64	–	-	–	-

The hydrogen thermal desorption analysis has shown the high binding energy of the rare earth metal associated trap sites, as indicated by high desorption peak temperature. It also shows the potential use of the rare earth metal additions to provide accelerated reduction of diffusible hydrogen during welding cooling cycle. However, evaluation of these trap sites needs to be further pursued to assess the kinetics aspects of hydrogen release and trapping of these trap sites. The binding energy only relates to the driving force of the hydrogen capturing, but not the rate.
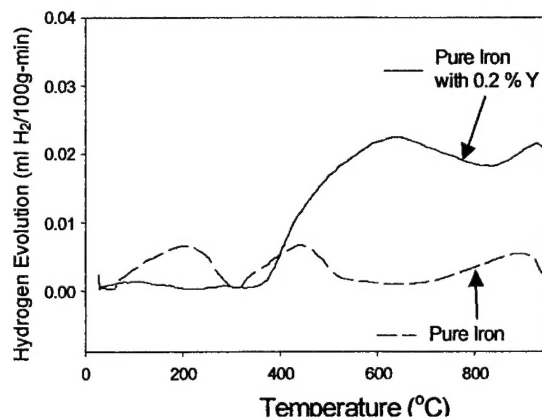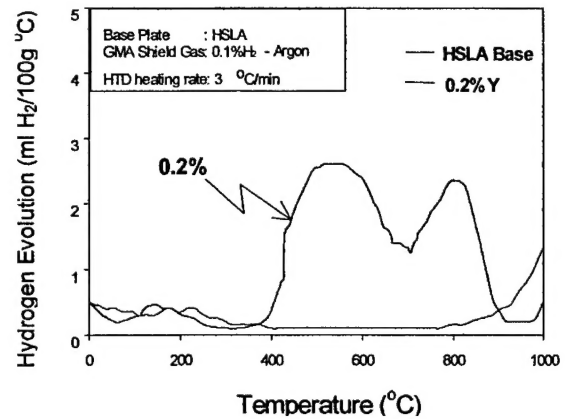
Fig. 5. Hydrogen thermal desorption of weld metal GMA welded with 0.5 pct. H_2 in argon shielding gas and 600 ppm addition in the weld metal. (a). High purity iron base metal (4 °C/min) (b). HSLA steel base metal (3 °C/min) [13].

II-2. Influence of Welding Process Parameters on the Hydrogen Trapping Effectiveness of Yttrium Addition

Yttrium ferro-additions have demonstrated the significant reduction in diffusible hydrogen content. Experiments were performed on pure iron and high strength low alloy steel weld metal and the proper yttrium content for its effective use has been determined. Also, the recovery of yttrium from the welding consumable to the weld pool has been measured for these experimental wires. The optimum set of welding process parameters for the effective use of the yttrium additions has been determined and demonstrated [16].

The addition of yttrium in the welding arc allows lower voltages and currents for the welding arc due to the lower ionization potential of yttrium than that of argon. These welds were made with 0.1 percent hydrogen in argon shield gas and the welding voltage at 30 volts. The effect on yttrium additions to the amount of heat, i.e. heat input, can be seen in figure 6. The heat inputs ranged from 1.3 to 2.7 kJ per millimeter. The heat input is an indication of the cooling rate that the weld pool will experience. At slower cooling rates, or larger heat inputs, more time is available for diffusion of hydrogen to be trapped by yttrium type traps and the effusion of hydrogen from the weld pool.

The effect of adding 650 ppm of yttrium was to increase the diffusible hydrogen content, as shown in Figure 6. This increase suggests that yttrium reacted with all of the available oxygen and the law of mass action for the oxygen-hydrogen reaction results in higher diffusible hydrogen content. With 3000 or 6000 ppm yttrium additions in the metal cored wired, there was sufficient yttrium to overcome the oxygen-hydrogen inverse relationship for the water reaction. The formation of the yttrium oxide inclusion is readily available for hydrogen trapping thus reducing the diffusible hydrogen content to low levels.

A metal transfer mode plot (Figure 7) was made for gas metal arc (GMA) welding process with a corresponding mapping of diffusible hydrogen contents. The ANSI/AWS A4.3-93 standard was used with gas chromatography to measure diffusible hydrogen contents of HSLA 100 steel welds. Metal cored steel wires with yttrium as a weld metal hydrogen trap were used. Depending on yttrium and sulfur contents yttrium oxide and/or yttrium oxy-sulfide inclusions form in the weld metal. The metal transfer modes were observed and recorded during welding and mapped accordingly with the welding voltage and current. Heat input ranged from 1.4 to 3 kJ/mm and shows a range less than one up to 4 ml of hydrogen per 100 g weld deposit. The effects of voltage, current, travel speed, polarity, and oxygen levels have also been examined. An optimum operating window is shown for welds made resulting in less than one ml of hydrogen (per 100 g deposit) content as shown in Figure 7. In the spray mode, yttrium is effectively transferred across the arc and has the necessary time during transfer across this arc for this size of droplet to produce sufficient yttrium compounds to serve as hydrogen trapping site.
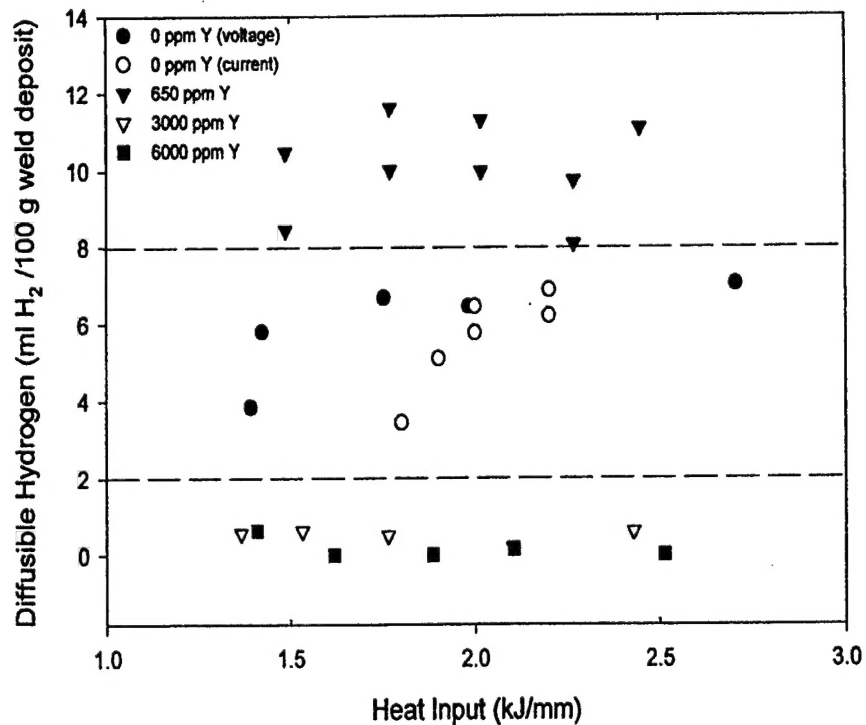
Figure. 6 The effect of heat input on the diffusible hydrogen content with yttrium hydrogen trap additions to metal cored steel wire. Welds made with 0.1 percent hydrogen in argon shield gas on high strength low alloy steel baseplate [16].
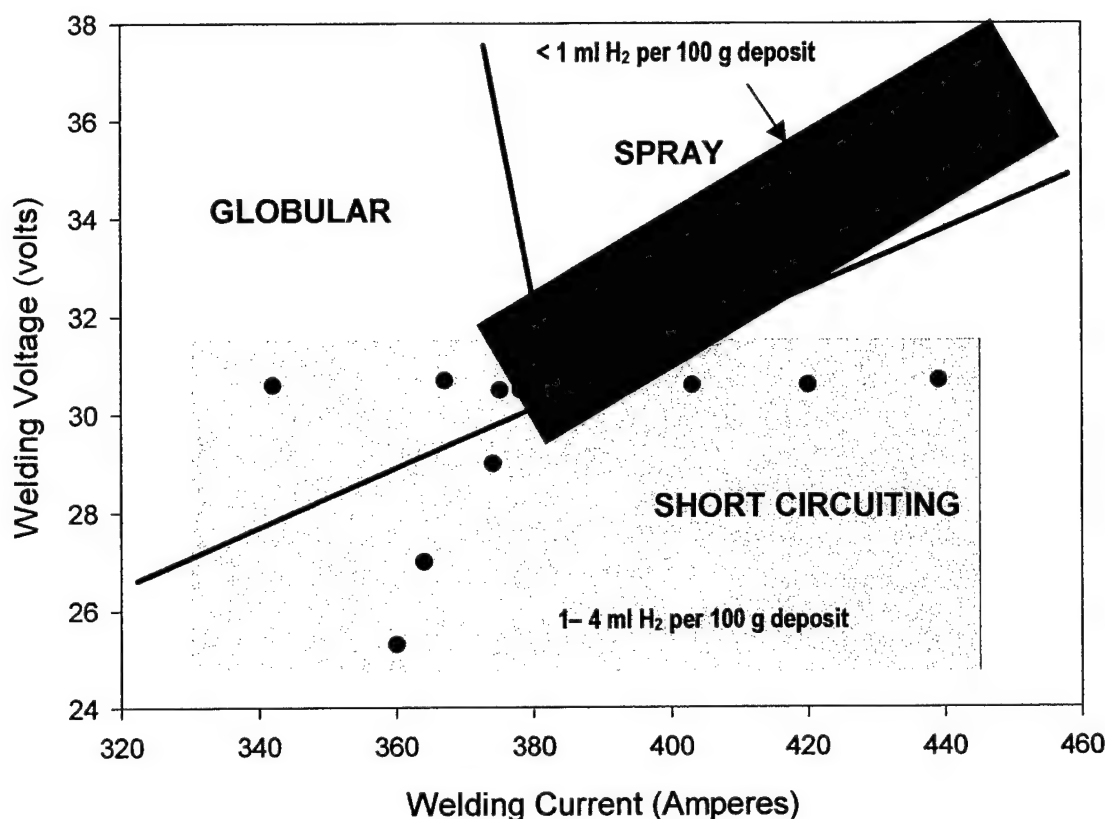
Figure 7. Metal transfer mode – weld metal diffusible hydrogen content plot on a GMA welded HSLA steel with HSLA metal cored steel wire containing 1600 ppm yttrium [16].

Thermal desorption curve for HSLA 100 steel welds made with yttrium additions to a HSLA steel metal cored wire with zero and four percent oxygen in a shielding gas of argon-0.1% hydrogen shows less trapped hydrogen for the four percent oxygen addition. The welds made with no oxygen additions have three peaks. Peak 1 is for a typical of microvoids (a reversible trap), Peak 2 is for a yttrium oxide trap (an irreversible trap), and Peak 3 is for yttrium-oxy-sulfide trap (an irreversible type trap). HSLA steel welds were made at 25 volts and with a wire feed speed of 240 ipm. High oxygen content shows a higher trapped (residual) hydrogen content in the weld metal and also the formation of primarily yttrium oxide inclusions. An increase in the hydrogen content suggests that increasing the shielding gas oxygen content significantly alters the size and distribution of weld metal inclusions and thus the available interfacial trap sites.
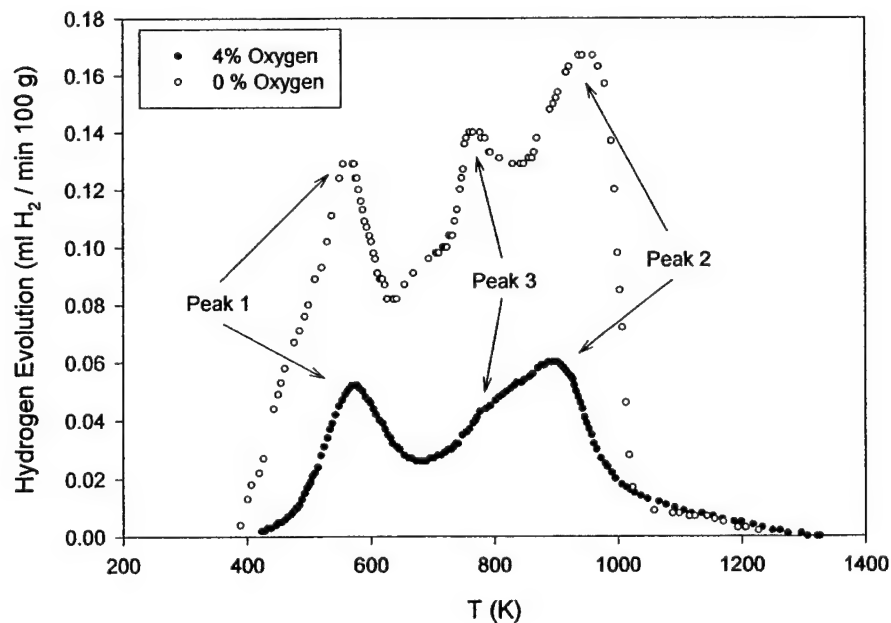
Figure 8. Thermal desorption curve for spray and globular transfer mode. Peak 1 is associated with microvoids; Peak 2 is yttrium oxy-sulfide, and Peak 3 as yttrium oxide [16].

III. HYDROGEN REDUCTION WITH FLUORIDE ADDITIONS

Hydrogen absorption can be minimized through the formation of water vapor or hydrogen fluoride in a thermo-chemical reaction with oxygen or fluorine, respectively, in the welding plasma. Increasing fluorine gas will decrease the amount of hydrogen available to enter the weld deposit as shown in Fig 9. These gases will be generated from selected fluxes that easily decompose during arc heating.

Fluorspar (CaF_2) is believed to dissociate at high temperatures to produce fluorine, and consequently shifts the reaction in Equation 1 to the right, resulting in reduced hydrogen in the arc.

$$F_2 + H_2 \leftrightarrows 2HF \qquad \text{(Eq. 1)}$$

However, it is also recognized that the decomposition of CaF_2 is not particularly active so that most of CaF_2 remains in the slag. Therefore, researchers have been searching for other fluorides more efficient in weld metal hydrogen reduction [17, 18].

Tsuboi et al. [19] observed a reduction of diffusible hydrogen with the addition of Na_3AlF_6 to the welding flux. At eight percent fluoride additions, the diffusible hydrogen level was 4 ml/100g of the deposited metal, which was thirty percent lower than that obtained using CaF_2. Pokhodnya et al. [18] also reported the effectiveness of complex fluorides such as Na_2SiF_6, Na_2TiF_6, K_2SiF_6, and K_2TiF_6 by using Flux Cored Arc Welding (FCAW). Similarly, Matsushita and Liu investigated diffusible hydrogen control in steel weld metal with fluoride additions in welding flux. Figure 10 illustrates the results of diffusible hydrogen

content in the steel weld metal deposited with experimental wires that contained 5% K_3AlF_6, 7.5% K_3AlF_6, and 10% K_3AlF_6 additions. The addition of K_3AlF_6 was found to be more effective than that of CaF_2 for hydrogen reduction [21]. Johnson and Liu [20] reported that MnF_3 was able to reduce diffusible hydrogen in welds performed on primer-coated steel plates.
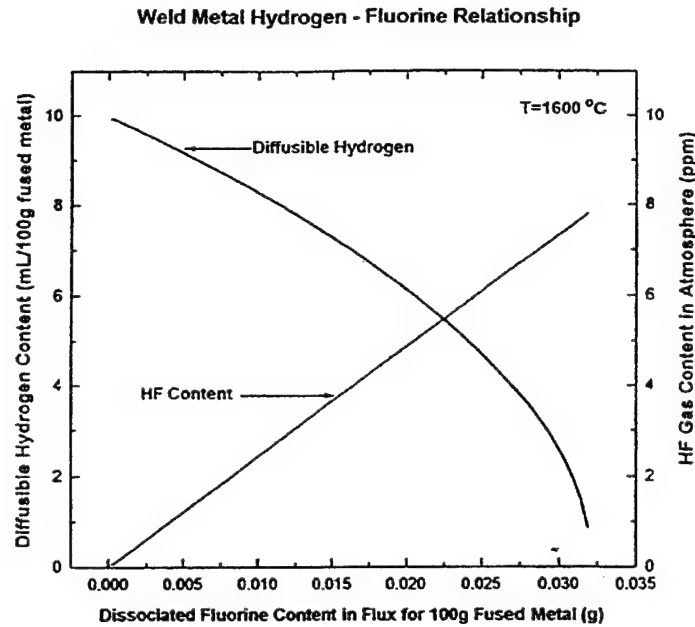
Weld Metal Hydrogen - Fluorine Relationship

Figure 9. Thermo-chemical reaction between fluorine and hydrogen in the welding plasma [2].
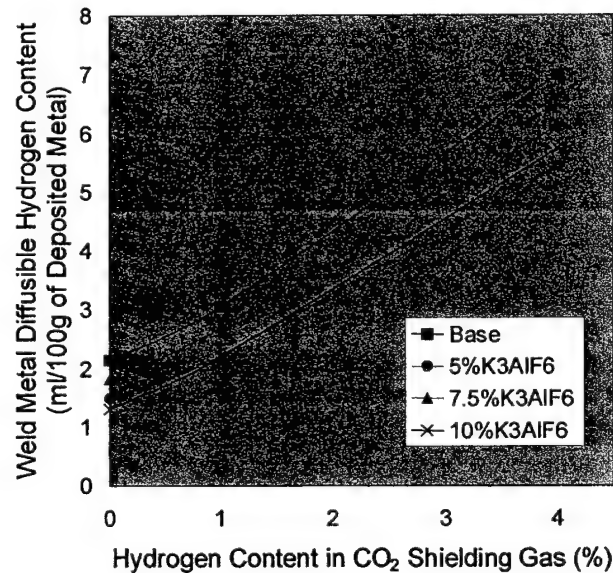
Figure 10. Steel weld metal diffusible hydrogen content given by Base 5% K_3AlF_6, 7.5 K_3AlF_6, and 10% K_3AlF_6 wires [21].

IV. ROLE OF RETAINED AUSTENITE IN HYDROGEN MANAGEMENT

Retained austenite was found to be an intraphase hydrogen trapping site which has relatively high binding energy. To identify retained austenite as an intraphase hydrogen trapping site, four samples with different austenite volume fraction were prepared: AISI type 304 stainless steel, super duplex stainless steel, dual phase steel (12 vol. pct. retained austenite), high strength low alloy (HSLA) steel weld deposit (4 vol. pct. and 0 vol. pct. retained austenite). Each of the samples was charged with 99.9 purity of hydrogen by using gaseous charging. The thermal desorption analysis was employed to observe the hydrogen trapping and detrapping behavior [22, 23, 24].

The hydrogen desorption of the four samples are shown in Figure 11. As clearly shown in this figure, all of the hydrogen desorption has a peak approximately at 600 °C. The corresponding amount of desorbed hydrogen decreased as the volume fraction of austenite decreased. The relationship between the amount of the desorbed hydrogen and the volume fraction of austenite is linear, as shown in Figure 12. These results led to a conclusion that retained austenite behaves as a strong hydrogen trap whose trapping capacity is directly proportional to its volume. Retained austenite is considered as a bulk hydrogen trap which may also contain more than one kind of hydrogen trap sites besides its lattice sites [22].

The exact nature of the interaction between hydrogen and retained austenite, which is manifested by such a high thermal desorption, is not clearly understood. It is worth noticing that there is a different situation of hydrogen desorption between the AISI 304 stainless steel sample and the HSLA steel weld sample. In the former sample, austenite remained stable during the course of thermal heating. Hydrogen seemed to be released from some type of trap sites, most likely dislocations and grain boundaries. Also, hydrogen degassed out of the sample by diffusing sluggishly through the stable austenite phase. In the HSLA steel sample, hydrogen may be released simply due to the progression of retained austenite dissolution during the course of thermal heating. Also in the HSLA steel sample, hydrogen transports more easily though the ferrite matrix. However its desorption was very similar to the AISI 304 stainless steel as seen by its similar peak temperature. An ongoing study is still being carried out to clarify the hydrogen-trapping characteristic of retained austenite [22].

Retained austenite is not only a diffusion-controlled trap but also a meta-stable phase. The dual phase steel (12 vol. pct. retained austenite) was investigated to observe the effects of reduced temperature by using thermal desorption analysis. When the reduced temperature were applied on hydrogen charged specimens, the thermal desorption analysis exhibited a shifting of hydrogen peak to lower temperature as shown in Figure 13(a). The measured retained austenite content as function of quenching temperature is shown Figure 13(b). This result suggests that microstructural transformation associated with changes in service conditions can result in a high diffusible hydrogen content in the resulting martensite phase, which is known as the most susceptible microstructure to hydrogen assisted cracking (HAC). Demonstration of the hydrogen trapping capacity and the metastability of retained austenite suggests that the amount of allowable retained austenite needs to be related to the amount of hydrogen pick up and the expected service condition [23].
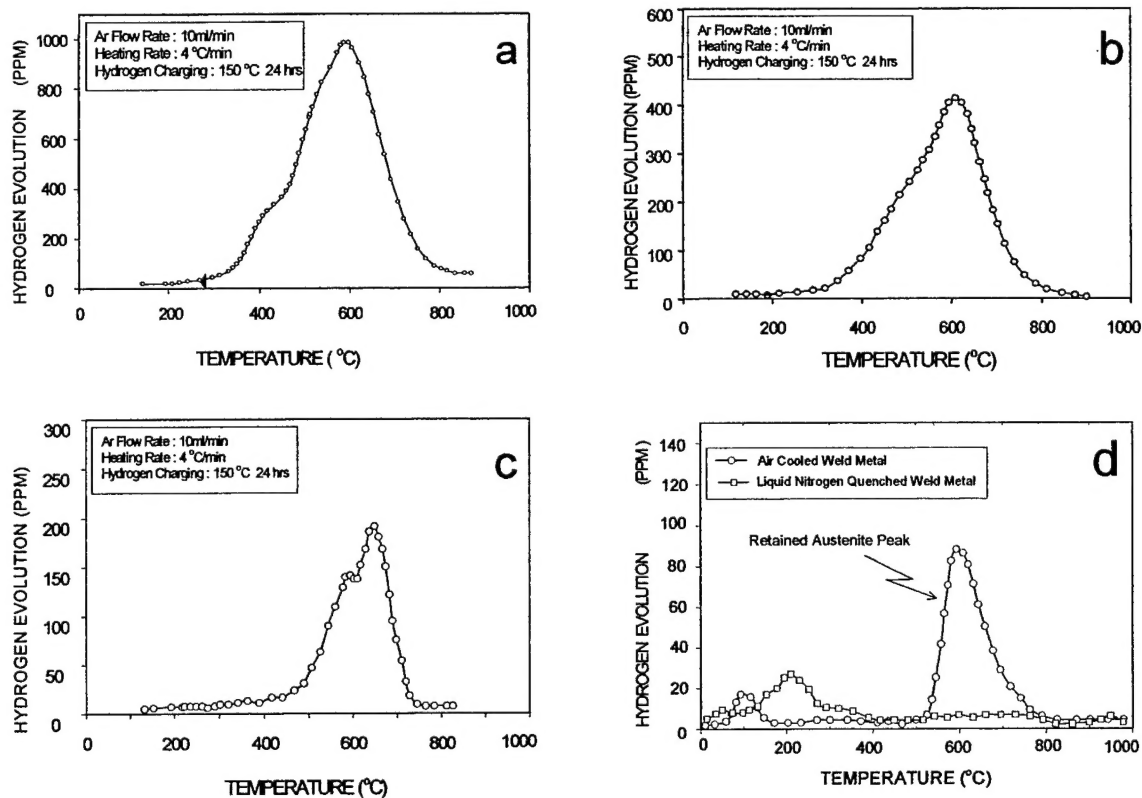
Fig. 11. Thermal desorption analysis for dual phase steel which is quenched in liquid nitrogen after charging; a-type 304 stainless steel b. superduplex stainless steel c. dual phase steel d. HSLA weld metal [22].
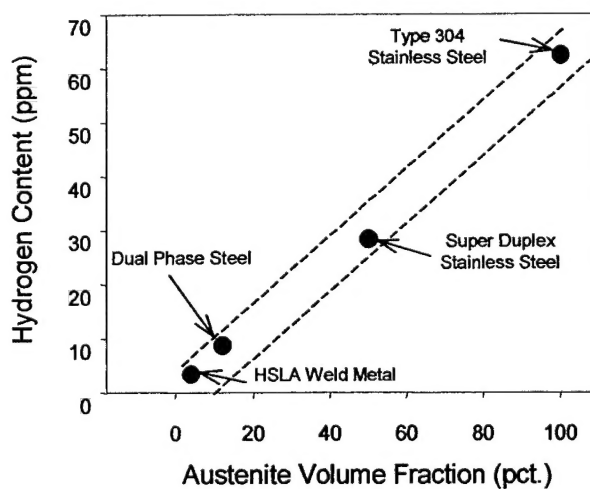

Fig. 12. The relationship between the amount of hydrogen release by desorption with the volume fraction of austenite (22).
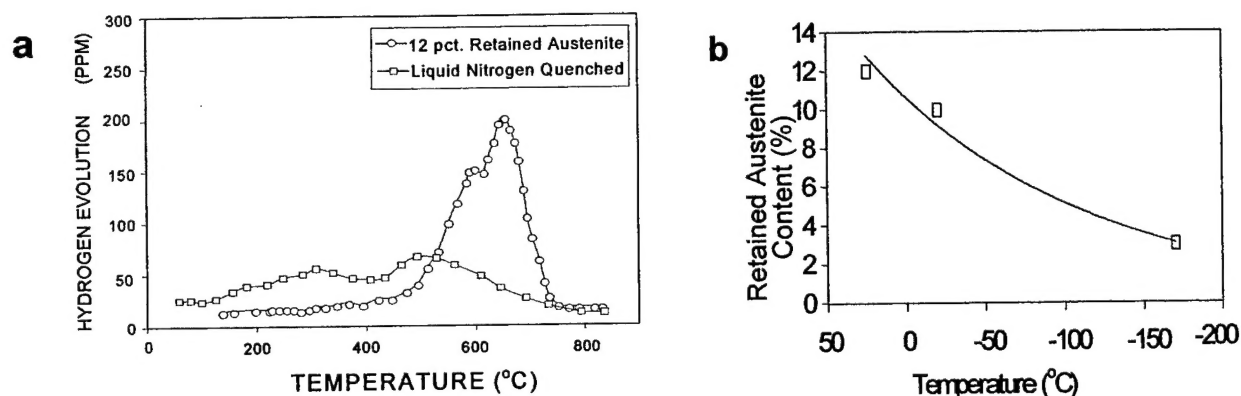
Figure 13. a-Thermal desorption analysis for dual phase steel which is quenched in liquid nitrogen after charging. b-The measured retained austenite content as function of quenching temperature [23].

SUMMARY

Diffusible hydrogen content in high strength low alloy steels welding can be better managed with modifications to the low hydrogen welding consumables. The welding consumable alloy content needs to allow the weld metal to transform from austenite to ferrite/martensite at slightly higher temperature than the base metal does. This practice will allow for the best dispersal of hydrogen from the weld deposit and minimizing the tendency of localizing the hydrogen in the heat affected zone at the fusion line. The use of weld metal hydrogen traps will both reduce the total diffusible hydrogen content and also minimize the hydrogen distribution near the fusion line. The use of complex metal fluorides in the welding consumable to achieve small but sufficient amounts of fluorine to the welding plasma can produce hydrogen fluoride, thus reducing the amount of hydrogen source available for the weld pool or the resulting the weld deposit. Also, the presence of retained austenite was found to influence the ability to control weld metal hydrogen in high strength low alloy steels.

CONCLUSIONS

1. Three modifications to low hydrogen welding consumables have been suggested to be use separately or all together to assist in achieving very low diffusible hydrogen contents in high strength low alloy steel welds.

2. Retained austenite in weld metal is a unpredictable hydrogen source, which upon austenite decomposition, due to thermal experiences may deliver unacceptable local amounts of hydrogen to hard phase transformation products and may resulting in increase of hydrogen cracking susceptibility.

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